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(54) Title: THERMALLY CONDUCTIVE GEL MATERIALS

(57) Abstract

A composition comprising a gel in which electrically non-conductive particulate filler is dispersed. The thermal conductivity of the particulate filler is at least 2 watts/m-°K. The composition has a thermal conductivity of at least 0.2 watts/m-°K and tack of at least 3g. Thermal management is improved by selecting a composition having properties which enable better adhesion between the composition and the surface away from which heat is to be conducted.

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THERMALLY CONDUCTIVE GEL MATERIALS

This invention relates to a thermally conductive composition and an assembly utilizing the composition.

Background of the Invention

Various compositions have been used to aid in the conduction of heat from one surface to another surface. For example, in electrical devices it is important to conduct heat away from electronic modules, enclosures, circuit boards and components toward a metal plate or cooling device which can aid in thermal management.

Various grease materials and pastes have been prepared using similar thermally conductive fillers. However, such greases and pastes have the disadvantages of migrating into adjacent spaces over time, particularly at elevated temperatures, thereby contaminating other areas of the device and causing a loss of the desired thermal conductivity. Greases and pastes are also very difficult to handle, particularly when re-entering the electrical device for repair or replacement, because they are difficult to clean from the surfaces on which they have been placed.

Elastomeric compositions have been used which contain various additives and fillers which increase the thermal conductivity of the elastomers. For example, U.S. Pat No. 4,852,646 issued on August 1, 1989 to Dittmer et al., which is commonly assigned with the present application and which is incorporated herein by reference, discloses a gel material including fillers for aiding in the conduction of heat from one surface to another. However, with vibration and thermal cycling, the interface between the substrates of the prior art may separate, and thermal transfer is impaired.

Summary of the Invention

We have now discovered that thermally conductive gels having a high tack provide remarkably good performance, especially in applications in which the thermally conductive material, during use, is subject to vibration and thermal cycling.

In a first aspect, this invention relates to an electrically insulating, thermally conductive, composition which

(1) comprises:

(a) a gel;

(b) dispersed in the gel, an electrically non-conductive particulate filler having a thermal conductivity of at least 2 watts/m-°K;

(2) has a thermal conductivity of at least 0.2 watts/m-°K;

(3) has a surface tension of 18 to 50 dyn/cm; and

(4) has a tack of at least 3 g.

In a second aspect, this invention relates to an assembly which comprises:

(1) a support; and

(2) secured to the support an electrically insulating composition which

(a) comprises:

(i) a gel;

(ii) dispersed in the gel, an electrically non-conductive particulate filler having a thermal conductivity of at least 2 watts/m-°K;

(b) has a thermal conductivity of at least 0.2 watts/m-°K;

(c) has a surface tension of 18 to 50 dyn/cm; and

(d) has a tack of at least 3 g.

In a further aspect, the invention relates to an assembly which comprises:

(1) a non-compressible support;

(2) secured to the support an electrically insulating composition which

(a) comprises:

(i) a gel;

(ii) dispersed in the gel, an electrically non-conductive particulate filler having a thermal conductivity of at least 2 watts/m-°K;

(b) has a thermal conductivity of at least 0.2 watts/m-°K;

(c) has a surface tension of 18 to 50 dyn/cm; and

(d) has a tack of at least 3 g;

(3) a substrate adjacent to the support; and

(4) an electrical component positioned on the substrate, extending through the support, and in contact with the composition.

Brief Description of the Drawing

The Figure is a cross sectional view of the assembly of the present invention.

Detailed Description of the Invention

The composition of the present invention comprises a gel in which electrically non-conductive particulate filler is dispersed. The composition is thermally conductive and electrically non-conductive and is most advantageously used for thermal management, sealing and shock absorption.

Thermal management is often required in printed circuit boards, cellular telephones, pagers, automotive junction blocks, and other electronic modules and enclosures which include contacts, resistors, diodes or other electrical components. The composition of the present invention adheres to the components, which improves conduction of heat away from the components.

Using known thermally conductive compositions, in order to ensure adequate adhesion, it is necessary to have compression of the assembly. The compositions of the present invention exhibit high tack, which provides an improved interface, even after extended use of the assembly, and can provide excellent thermal conductivity, even with little or no compression.

The composition of the present invention is preferably a gel obtained by blending at least one prepolymer with an extender and a particulate filler, and then subjecting the blend to conditions which convert the prepolymer into gel. The gel is a substantially dilute crosslinked system which exhibits no flow when in the steady-state. The crosslinks, which provide a continuous network structure, may be the result of physical or chemical bonds, crystallites or other junctions, and must remain intact under the use conditions of the gel. Most gels comprise a fluid-extended polymer in which a fluid, e.g., an oil, fills the interstices of the network.

Gels useful in the present invention include those comprising silicone, for example, a polyorganosiloxane system, polyurethane, polyurea, styrene-butadiene copolymers, styrene isoprene copolymers, styrene-(ethylene/propylene)-styrene (SEPS) block copolymers (available under the tradename Septon™ by Kuraray), styrene-(ethylene-propylene/ethylene-butylene)-styrene block copolymers (available under the tradename Septon™ by Kuraray), and/or styrene-

(ethylene/butylene)-styrene (SEBS) block copolymers (available under the tradename Kraton™ by Shell Oil Co.).

Suitable extender fluids include mineral oil, vegetable oil, paraffinic oil, silicone oil, placticizer such as trimellitate, or a mixture of these, generally in an amount of 30 to 90% by weight, based on the total weight of the gel. The gel may be a thermosetting gel, for example silicone gel, in which the crosslinks are formed through the use of multifunctional crosslinking agents, or a thermoplastic gel, in which microphase separation of domains serves as junction points. Disclosures of gels which may be suitable as the polymeric component in the composition are found in U.S. Patent Nos. 4,600,261 to Debbaut, 4,690,831 to Uken et al, 4,716,183 to Gamarra et al, 4,777,063 to Dubrow et al, 4,864,725 to Debbaut et al, 4,865,905 to Uken, 5,079,300 to Dubrow et al, 5,104,930 to Rinde et al, and 5,149,736 to Gamarra, and in International Patent Publication Nos. WO 86/01634 to Toy et al, WO 88/00603 to Francis et al, WO 90/05166 to Sutherland, WO 91/05014 to Sutherland, and WO 93/23472 to Hammond et al. The disclosure of each of these patents and publications is incorporated herein by reference.

The prepolymer preferably has a viscosity of 1,000 to 8,000 CPS at room temperature, more preferably 2,000 to 6,000 CPS, particularly 2,000 to 4,000 CPS, and especially 2,500 to 3,500 CPS. The extender preferably has a viscosity of 1,000 to 8,000 CPS at room temperature, more preferably 2,000 to 6,000 CPS, particularly 2,000 to 4,000 CPS, and especially 2,500 to 3,500 CPS. The viscosity of the prepolymer and the extender is an important factor in determining tack of the final gel composition.

The particulate filler has a thermal conductivity of at least 2 watts/m-°K, preferably 2 to 100 watts/m-°K, particularly 10 to 20 watts/m-°K. The particulate filler is preferably present in amount at least 25%, more preferably 40 to 65%, particularly 60 to 70%, by weight, based on the weight of the composition. The particulate filler materials useful in the present invention are electrically non-conductive so as to prevent short circuits with electrical components. Particularly preferred fillers are one or more of alumina, zinc oxide, aluminum nitride, boron nitride, synthetic diamond, magnesium and magnesium oxide. The filler may be of any physical shape and form desired to provide the thermal conductivity and retain the desired properties of the composition. For example, the particulate fillers may be powders of varying particle sizes and of any desired shape, such as round, irregular, flake or platelet type particles, cubic, hexagonal or other shape, depending upon the processing used to prepare the particulate filler. In some cases, it may be desired to use a particulate filler which is in the form of short fibers such as chopped fibers or in the form of short needles or whiskers provided that the

length and stiffness thereof do not interfere with the conformability of the composition to the shape of the surface on which the composition is intended to be used for aiding in heat transfer. The particulate materials useful in the gel compositions of this invention can also be preformed into matrix forms such as woven, nonwoven, mat, sheet, or other form provided that the preformed particulate material is porous enough for sufficient impregnation by the liquid gel material before it is cured and provided that the preformed particulate material is sufficiently flexible to conform as desired as part of the gel composition of this invention.

In addition to the particulate filler, the composition may include other conventional additives, including stabilizers, pigments, crosslinking agents, catalysts and inhibitors.

The final composition has tack of at least 3 g, preferably 3 to 40 g, particularly 8 to 20 g; thermal conductivity of at least 0.2 watts/m-°K, preferably 0.2 to 1.0 watts/m-°K, particularly 0.2 to 0.8 watts/m-°K, especially 0.4 to 0.6 watts/m-°K; preferably has surface tension of 18 to 50 dyn/cm, more preferably 18 to 26 dyn/cm, particularly 20 to 22 dyn/cm; preferably has a Voland hardness of 12 to 200 g, more preferably of 40 to 150 g. The Voland hardness and tack are measured using a Voland-Stevens Texture Analyzer Model LFRA having a 1000 g load cell, a 5 g trigger, and a 0.25 inch (6.35 mm) ball probe, as described in U.S. Patent No. 5,079,300 to Dubrow et al, the disclosure of which is referred to above. To measure the hardness of a gel, a 20 ml glass scintillating vial containing 10 g of gel is placed in the analyzer and the stainless steel ball probe is forced into the gel at a speed of 0.20 mm/second to a penetration distance of 4.0 mm. The Voland hardness value is the force in grams required to force the ball probe at that speed to penetrate or deform the surface of the gel the specified 4.0 mm. The Voland hardness of a particular gel may be directly correlated to ASTM D217 cone penetration hardness using the procedure described in U.S. Patent No. 4,852,646 to Dittmer et al, referred to above.

An assembly according to the present invention includes the composition of the present invention, as described above, secured to a support. The support can be composed of a polymeric material which is non-compressible, more preferably a polyester such as polyethylene terephthalate, or polypropylene. For example, the composition may overlie a polypropylene support, or may be sandwiched between two thin, laminar polyester supports.

In a preferred embodiment, the composition is reinforced internally by a sheet, preferably a flexible matrix. The matrix serves to improve the mechanical properties of the composition, such as tensile strength and modulus of elasticity. The composition may be impregnated in a reinforcing sheet, for example, a film or matrix constructed of foam or fabric.

A fabric matrix may be a single layer or a plurality of layers. A preferred material for the flexible matrix is polyurethane. Gel impregnated in a matrix is disclosed in U.S. Patent No. 4,865,905 to Uken, referred to above.

The assembly may be a solid sheet, or may have cut-outs formed therein to adhere to components, thereby aiding in thermal transfer.

In use, the assembly is positioned in contact with surfaces to aid in the conduction of heat away from the source. Because the composition has high tack, i.e., at least 3 g, preferably 3 to 40 g, and particularly 8 to 20 g, it adheres strongly to the surfaces and electrical components, even, over time, with vibration and thermal cycling.

Referring to the Figure, an assembly 2 includes a composition 4, including a gel having particulate filler 6 dispersed therein. Composition layer 4 is greater than 1 mm thick and is sandwiched between non-compressible, laminar supports 8 constructed of polyester, in this case, polyethylene terephthalate. Supports 8 each have a thickness of 1 mm. Composition layer 4 is reinforced by a flexible matrix 10, composed of a polyurethane.

Assembly 2 is placed on a substrate 12, which includes an electrical component 14 positioned on the substrate. Electrical component 14 is in close contact with assembly 2 and is in contact with and adheres to composition 4.

Example 1

A sample was prepared by mixing a commercially available two part addition cure polydimethylsiloxane gel. Each part has a starting viscosity of 3000 CPS before gelation. Part A, which contains a polydimethylsiloxane, and Part B, which contains an extender and a crosslinking agent, were mixed together along with 25% by weight, based on the total weight of the composition, of Al_2O_3 in the form of a fine powder having a thermal conductivity of 15 watts/m-°K, a diameter of 3 to 4 μm, a density of 4 g/cc and 325 mesh (available from Cerac as 1005). These components were mixed in a container. A strip of flexible matrix was continuously passed through the container and impregnated with the mixture. The sample was then flash cured in an oven at 150°C for 1 minutes and wound around a take-up reel. The sample was then post cured at 80°C for 12 hours. The cured composition had a Voland hardness of 29 g, tack of 10 g, surface tension of 22 dyn/cm, and thermal conductivity of 0.35 watts/m-°K.

Example 2

Example 1 was repeated, but using 50% of Al_2O_3. The cured composition had a Voland hardness of 112 g, tack of 10 g, surface tension of 22 dyn/cm, and thermal conductivity of 0.5 watts/m-°K.

What is Claimed is:

1. An electrically insulating composition which
   (1) comprises:
      (a) a gel;
      (b) dispersed in the gel, an electrically non-conductive particulate filler having a thermal conductivity of at least 2 watts/m-°K;
   (2) has a thermal conductivity of at least 0.2 watts/m-°K;
   (3) has a surface tension of 18 to 50 dyn/cm; and
   (4) has a tack of at least 3 g.

2. The composition of claim 1 wherein the composition is one obtained by
   (1) blending
      (a) at least one prepolymer having a viscosity of 1,000 to 8,000 CPS at room temperature; and
      (b) an extender having a viscosity of 1,000 to 8,000 CPS at room temperature; and
      (c) the filler; and
   (2) subjecting the blend to conditions which convert the prepolymer into a gel.

3. The composition as defined in any one of the preceding claims which has a thermal conductivity of 0.2 to 1.0 watts/m-°K.

4. The composition as defined in any one of the preceding claims which has a tack of 8g to 20g.

5. The composition as defined in any one of the preceding claims wherein the gel is a polyorganosiloxane.

6. The composition as defined in any one of the preceding claims which has a Voland hardness of 12 to 200 g.

7. The composition as defined in any one of the preceding claims wherein the filler is present in amount of at least 25% by weight, based on the weight of the composition.

8. The composition of claim 7 wherein the filler is present in amount of 40 to 70% by weight, based on the weight of the composition.

9. The composition of claim 1 wherein the filler is one or more of alumina, zinc oxide, aluminum nitride, boron nitride, synthetic diamond and magnesium oxide.

10. The composition of claim 9 wherein the filler consists essentially of alumina.

11. An assembly which comprises:
    (1) a support; and
    (2) secured to the support an electrically insulating composition which
        (a) comprises:
            (i) a gel;
            (ii) dispersed in the gel, an electrically non-conductive particulate filler having a thermal conductivity of at least 2 watts/m-°K;
        (b) has a thermal conductivity of at least 0.2 watts/m-°K;
        (c) has a surface tension of 18 to 50 dyn/cm;
        (d) has a tack of at least 3 g.

12. The assembly of claim 11 which has a tack of 8g to 20g.

13. The assembly as defined in claim 11 or 12 wherein the filler is present in amount of 40 to 70% by weight, based on the weight of the composition.

14. The assembly as defined in claim 11, 12 or 13 wherein the support is non-compressible.

15. The assembly as defined in claim 11, 12, 13 or 14 wherein the support is composed of a polyester.

16. The assembly of claim 15 wherein the polyester is polyethylene terephthalate.

17. The assembly as defined in claim 11, 12, 13, 14 or 15 wherein the support is composed of polypropylene.

18. The assembly as defined in claim 11, 12, 13, 14, 15, 16 or 17 wherein the composition is sandwiched between two laminar supports.

19. The assembly of as defined in claim 11, 12, 13, 14, 15, 16, 17 or 18 wherein the composition is internally reinforced by a flexible matrix .

20. An assembly which comprises:

(1) a non-compressible support;

(2) secured to the support an electrically insulating composition which

(a) comprises:

(i) a gel;

(ii) dispersed in the gel, an electrically non-conductive particulate filler having a thermal conductivity of at least 2 watts/m-°K;

(b) has a thermal conductivity of at least 0.2 watts/m-°K;

(c) has a surface tension of 18 to 50 dyn/cm; and

(d) has a tack of at least 3 g;

(3) a substrate adjacent to the support; and

(4) an electrical component positioned on the substrate, extending through the support, and in contact with the composition.

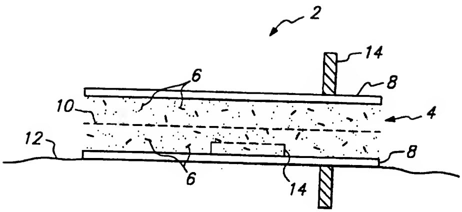

FIG.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 95/10209

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 H01B3/46 H01L23/24 H01L23/42 C08K3/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 H01B H01L C08K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DATABASE WPI Derwent Publications Ltd., London, GB; AN 94-216382 & TW,A,224 541 (SIX PHASE CO. LTD.) , 1 June 1994 see abstract ---	1,11
X	EP,A,0 603 928 (DELCO ELECTRONICS CORP) 29 June 1994 see claims 1,6,12 see page 2, line 50 - line 51 see page 3, line 21 - line 32 see page 3, line 33 - line 40 see figure 1 --- -/--	1,3

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
12 December 1995	29. 12. 95

Name and mailing address of the ISA	Authorized officer
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INTERNATIONAL SEARCH REPORT

Inter 'onal Application No
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